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Development of a natural analogue database to support the safety case of the Korean radioactive waste disposal program

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Abstract In this study, the status of natural analogue studies in Korea is briefly summarized and applicability of existing natural analogue information to the Korean safety case has been evaluated. To enable effective application of natural analogue information to the overall evaluation of long-term safety (the "safety case") for the geological disposal of radioactive wastes, a natural analogue database has been developed by collecting, classifying, and evaluating relevant data. The natural analogue data collected were classified into categories based on site information, components/processes of the disposal system, properties/phenomena, reference, safety case application, application method, and suitability to a safety case. Suitability of the natural analogue data to a specific safety case was evaluated based upon the importance and the applicability to the Korean safety case. As a result, 75 natural analogue datasets were selected as important for the Korean safety case. The database developed can now be utilized in the RD&D (Research, Development, and Demonstration) program development for natural analogue studies. In addition, the methodology developed and the database compiled in this study may assist in the development of safety case including safety assessment for high-level radioactive waste disposal in Korea as well as in other countries.

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1 Introduction

Plans for disposal of radioactive wastes in deep geological repositories have raised a number of issues, and presented great challenges, owing to the long geologic timescales over which such repositories must function. One possible way to improve long-term performance and safety assessments of a radioactive waste repository is to carry out natural analogue studies (Papp 1987; IAEA 1989, 1999; Smellie et al. 1997; Miller et al. 2000). Natural analogue studies have been carried out for more than three decades, although the scientific application of natural analogue information has only recently become well-organized (IAEA 1999). An analogue study of natural geological and geoarchaeological systems is useful in understanding the complexity of repositories over long periods of time, and can provide qualitative and quantitative data for safety assessment. Analogue studies are now considered to be an integral part of most national programs for the geological disposal of radioactive waste (Smellie et al. 1997). In addition, natural analogues can be used as complementary indicators for the development and illustration of the safety of a radioactive waste disposal system (Posiva 2012).

In Korea, KAERI (the Korea Atomic Energy Research Institute) has proposed a conceptual design of a geological disposal system for the direct disposal of spent nuclear fuel from PWR (Pressurized Water Reactor) and CANDU-PHWR (Canadian Deuterium Uranium Reactor—Pressurized Heavy Water Reactor) (Choi et al. 2013). Officials in the Korean government launched a project to develop an

advanced fuel cycle, based on the pyroprocessing of PWR spent fuel, to reduce the amount of high-level radioactive waste and to reuse the valuable fissile material. Consequently, KAERI has been developing a geological disposal system for high-level wastes from the pyroprocessing of PWR spent fuel since 2007 (Choi et al. 2013).

The plan for managing the high-level radioactive waste generated from the pyroprocessing is to dispose of the wastes in a deep crystalline geological formation such as granite (Choi et al. 2011). The design of the disposal system, and performance assessment, has been based on geological data collected from KURT (the KAERI Underground Research Tunnel), a facility in the KAERI research area, Daejeon city, in the middle of South Korea (Cho et al. 2008). The disposal canister, designed for waste blocks and storage canisters, is a copper-nodular, cast iron container with a predicted lifetime of 1000 years. The outer copper shell is 1.0 cm thick, and will be manufactured using a cold spray technique (Choi et al. 2013). Two disposal options are herein proposed and compared: vertical deposition and horizontal deposition. Korean Ca-bentonite with a dry density of 1.6 g/cm^3 is proposed as a buffer material.

In recent years, the scope of the safety assessment has broadened to include the collation of a broad range of evidence and arguments that complement and support the reliability of the results of quantitative analyses and the broader term "safety case" is used to refer to these extended studies (NEA 2004). A safety case is the synthesis of evidence, analyses and arguments that quantify and substantiate a claim that the repository will safe after closure and beyond the time when active control of the facility can be relied on (NEA 2004; see Reijonen et al. 2015, this issue). Natural analogues can support such safety case by providing evidence necessary for the long-term performance of a repository. Natural analogues have also proved to be essential and useful in the safety assessment of a repository. Furthermore, natural analogues can play important roles in public relations and education, which are very important issues in the safety case.

As geological disposal systems have developed in Korea, KAERI has carried out safety assessments for them based on the classical safety assessment concept (Lee and Hwang 2009; Hwang and Kang 2010). Recently, officials at KINS (the Korea Institute of Nuclear Safety), the nuclear regulatory body, made plans to apply the safety case concept in the licensing of radioactive waste repositories, especially for high-level radioactive waste repositories. Thus, KAERI has developed a safety case for the geological disposal of high-level radioactive waste in Korea. KINS also announced that natural analogues should be included in the development of the safety case for high-level radioactive waste disposal as a supplementary safety indicator. Consequently, KINS is planning to include this announcement in a new general regulation for high-level radioactive waste disposal in Korea. For these reasons, the need for natural analogue studies has recently increased for the development of the safety case in Korea.

Nevertheless, it remains a challenge to use natural analogue information appropriately in a safety case. A matrix system of natural analogue information, for example, could be a good approach for its application to the safety case (Miller et al. 2006). However, methodologies for the application of natural analogue information to the safety case have not been well developed, and proposed methodologies are few. Thus, it is essential to develop an easily-used methodology. In this study, a brief overview of the research status of natural analogue studies in Korea is presented. Progress in development of a natural analogue database for effective use in the development of the safety case for radioactive waste disposal, particularly suitable for high-level radioactive waste disposal in Korea, is reported.

2 Status of natural analogue studies in Korea

Only a few natural analogue studies have been carried out in Korea, in relation to radioactive waste disposal. From 1996 to 1999, KAERI participated in a project called Analogue Studies in the Alligator Rivers Region (ASARR) which was focused on the Koongarra uranium ore bodies, in the Alligator Rivers region of Australia (Payne and Airley 2006). In the ASARR project, KAERI developed a thermodynamic sorption model for application to natural composite materials (e.g., soil samples) taken from the Koongarra uranium deposit (Jung et al. 1999). A reactive flow and transport model was also developed which incorporated chemical reactions, recoil processes, and phase transformation of minerals, using a mixing-cell concept (Keum and Hahn 2003).

Some investigations of uranium ore deposits in Korea have been carried out for the development of uranium mining. However, actual mining has not taken place because the uranium grade is not economical. The largest, good-quality deposits are the Okchon group of uranium deposits located in the Okchon fold belt in the middle of South Korea. Most of the geological formations of the Okchon uranium deposits are metasedimentary rocks, such as uraniferous black slate, perlite, phyllite and chlorite schist, dolomite, and limestone. In a feasibility study, a uranium deposit in the Deok-pyoung area, located in the Goesan municipality within the Okchon fold belt, was proposed as a candidate site for natural analogue research, but no study has yet been started. However, some studies have been carried out using rock samples from KURT (Baik et al. 2004; Lee and Baik 2009). The main rock of

KURT is moderately fractured two-mica granite, composed mainly of quartz, microcline, biotite, muscovite, and plagioclase (Cho et al. 2008). Figure 1 shows a photograph of KURT, including some research works. The distribution of uranium in some rock samples from KURT was investigated for fresh intact rock, iron-coated rock, and fault minerals (Baik et al. 2004). The effect of weathering on uranium sorption onto granite has also been investigated (Lee and Baik 2009). Analogue studies for geochemical behavior of uranium in groundwater are now being carried out by analyzing uranium isotopes and speciation in KURT groundwater.

3 Natural analogue information database

3.1 Construction of a natural analogue information database

Various methods have been developed for the application of natural analogue information to the safety case for the geological disposal of radioactive wastes. The methods are generally classified into direct and indirect methods. The direct methods include quantitative, qualitative, and illustrative methods and the indirect methods include identification of FEPs (features, events, and processes) and scenario development (Smellie et al. 1997). Figure 2 shows the relationship between natural analogue information and safety case components (for instance, qualitative and quantitative data) (Miller et al. 2000). The qualitative data include model construction/development, and the quantitative data include data acquisition and model calibration/validation. These further expand to include non-technical illustration, performance assessment, and site characterization.

In the study reported here, a natural analogue database has been developed using a spreadsheet. For the construction of this database, 176 datasets were collected from various papers, reports, and books. The collected natural analogue datasets were categorized and evaluated for their applicability to a specific safety case (i.e., the Korean safety case). The main categories considered in the database are shown as part of a spreadsheet in Fig. 3. The categories in the spreadsheet are:

1. Geological formation: geological formation of a considered natural analogue site.
2. Natural analogue site: the name of the natural analogue site considered.
3. Nationality: the nationality of the natural analogue site considered.
4. System: barrier system of a repository such as engineered barrier system (EBS) and host rock.
5. Components/processes: components or processes of the EBS and host rock, respectively.
6. Properties/phenomena: properties or phenomena related to the component or process considered.
7. Basic information: information that can be obtained (e.g., hydraulic barrier, colloid filtration).

Fig. 1 A photograph of KURT (KAERI Underground Research Tunnel) which shows the portal (*background*), a hydraulic test (*lower left*), and a solute migration facility (*lower right*)

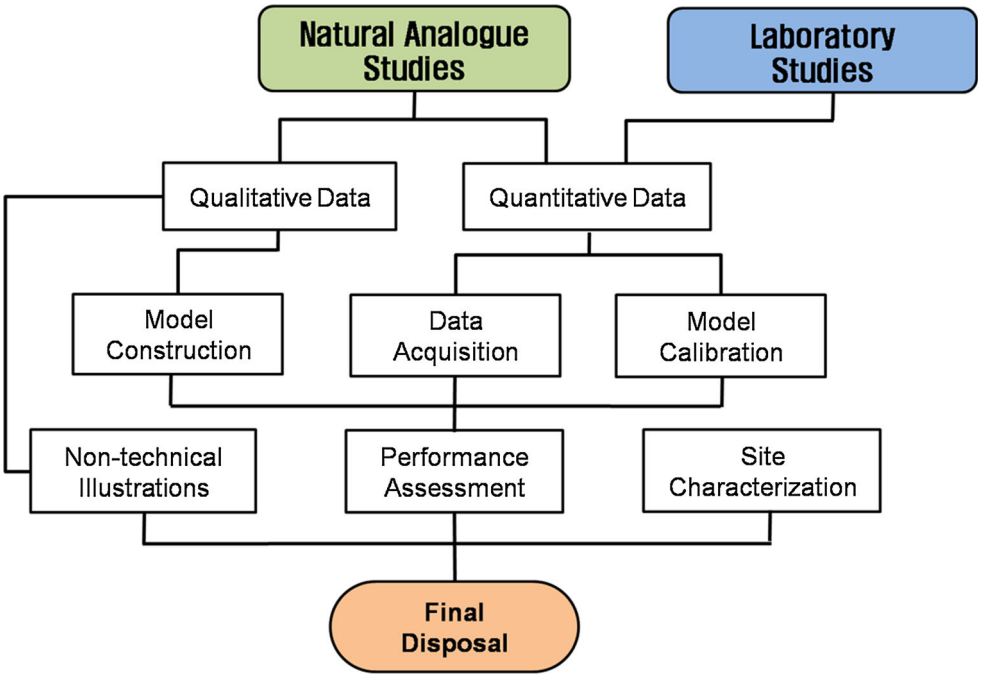


Fig. 2 Relationship between safety case components and natural analogue information (after Miller et al. 2000)

8. Natural analogue information: brief description of the natural analogue information.
9. Reference: the main reference(s) from which the natural analogue information was obtained.
10. Safety case: the safety case that the natural analogue information was applied to, if any.
11. Application: classification of the natural analogue information according to the methods used for application to the safety case.
12. Suitability: evaluation of the natural analogue information based upon the suitability to a specific safety case (the case of South Korea).

NO	Geological formatic	NA Site	Nationality	System	Components /Processes	Properties /Phenomena	Basic Information	NA information	Reference	Safety case	Application	Suitability
1	Granite	El Berrocal	Spain	Host rock	Matrix diffusion	Depth and volume of interconnected porosity	Diffusion depth	• Matrix diffusion limited to the first few tens of millimeters of rock adjacent to the fracture surface but that, within the rock matrix, the mobilized U was associated with secondary phases and is located in thin micro fissures and along grain boundaries.	Heath, 1995		UI	1
2	Granite	El Berrocal	Spain	Host rock	Elemental retardation	Transport and retardation within fractured crystalline rocks	Fracture mineralogy and precipitation	• Fracture filling mineral, adsorption and/or precipitation - Fracture minerals consist of quartz, potassium, feldspar, clay minerals, carbonates and minor pyrite and iron oxyhydroxides. U was mainly precipitate as mineral phases or coprecipitated with calcite - Fracture fillings are enriched in U by a factor ranging between 60 and 2, and in Th by a factor of 3.	Rivas et al., 1997; Perez del Villar et al., 1997		UI	1
3	Granite	Palmottu	Finland	Host rock	Matrix diffusion	Depth and volume of interconnected porosity	RN diffusion & depth	• Investigation on disequilibrium in 238U-234U-230Th - Distribution of radionuclides decreasing from fracture surface into the rock in an almost exponential fashion, diffuse alpha 80 mm into rock matrix; only RN loosely bound to the iron-rich phases diffused, channeled fluid flow even at the microscopic level, diffusion was predominantly along grain boundaries, - Simplistic conceptual diffusion model was tested as a "0th order" numerical interpretation of the U profiles.	Suksi & Ruskeeniemi, 1992		ISP, MV	1
4	Sediment	Dunarobba	Italy	EBS	Bentonite	Hydraulic barrier and colloid filter functions	Hydraulic and microbial activity barrier	• 2 million year old preserved forest (enveloped in lacustrine clay, above which are sand deposits with freely circulating, oxidizing water, clay envelope restrict ingress of the oxygenated water)	Ambrosetti et al., 1992; Benvegnu et al., 1988)	SR-97 (Sweden, 1999)	DE	1
	①	②	③	④	⑤	⑥	⑦	⑧	⑨	⑩	⑪	⑫

① Geological formation
② NA site
③ Nationality
④ System
⑤ Components or Processes
⑥ Properties or Phenomena
⑦ Basic information
⑧ NA Information
⑨ References
⑩ Safety case
⑪ Application
⑫ Suitability

Fig. 3 Sample of the developed natural analogue (NA) information database using a spreadsheet with the considered categories

Table 1 Elements of the categories for the system, components/processes, and properties/phenomena considered in the natural analogue database

System	Components/processes	Properties/Phenomena
EBS	Silicate glass	Devitirification
		Radiation induced effects
		Dissolution and alteration
		Radionuclide retardation by secondary alteration products
	Spent fuel	Dissolution and radionuclide release
		Radionuclide retardation by secondary alteration products
	Metal	Durability and longevity of iron and steel
		Durability and longevity of copper
		Radionuclide retardation by secondary alteration products
	Bentonite	Longevity of bentonite and the rate of alteration
		Physico-chemical changes due to heating
		Canister sinking
		Interaction with other repository materials
		Hydraulic barrier and colloid filter functions
	Concrete and cement	Durability of cement
		Cement-rock-groundwater interactions
		Radionuclide sorption
		Colloid production and filtration
		Gas and water permeability
		Bonding properties of cement and concrete
	Bitumen	Durability and longevity
		Groundwater leaching
		Microbial degradation
		Radiation induced degradation
		Interaction with saline water
	Organic material	Cellulose degradation
		Cellulose degradation products
		Natural resins
	Elemental solubility and speciation	Elemental solubility
		Speciation
Host rock	Elemental solubility and speciation	Elemental solubility
		Speciation
	Elemental retardation	Transport and retardation within fractured crystalline rocks
		Transport and retardation within argillaceous rock
		Transport and retardation at the geosphere-biosphere interface
		Measurement of in situ distribution coefficient
	Matrix diffusion	Depth and volume of interconnected porosity
		Bulk rock chemical buffering capacity
		Extent of matrix diffusion in sedimentary formation
		Estimation of diffusion coefficient
	Radiolysis	Process involved in radiolysis of groundwater
		Radiolysis in nature (how common is radiolysis in nature)
		Potential buffering capacity of reduced iron corrosion phases
	Redox front	Redox front formation and behavior in crystalline rocks
		Redox front formation and behaviour in argillaceous rocks
		Radionuclide migration at a redox front
	Colloids	Populations of colloids in natural systems
		Stability of colloids in natural systems
		Radionuclide uptake and transport by colloids in natural system
		Colloid in anthropogenic systems
		Biocolloids
	Microbial activity	Microbial populations at depth in natural systems
		Tolerance to hyper alkaline conditions
		Nutrient and energy availability
	Gas generation and migration	Gas production rates
		Gas migration and reaction with the geosphere
		Gas migration effects on solute transport

EBS engineered barrier system

Table 1 shows the elements of the categories for the EBS and the host rock systems (e.g., the "component/process" and "properties/phenomena" items in the spreadsheet) considered in the natural analogue database. If necessary, more elements can be added to the database because there could be more elements of the categories than those listed in Table 1.

3.2 Methods for application to the safety case

All the natural analogue datasets collected were classified into five categories according to the methods used to apply the natural analogue information to the safety case (Table 2). Quantitative methods include ISP (Input for Safety and Performance assessments) and MV (Model Validation). Qualitative methods included SD (Scenario Development

Table 2 Categories of the application methods of natural analogue datasets used in the database

Method	Categories (code)	Description
Quantitative	Input for safety and performance assessments (ISP)	Direct application as input parameters for safety/performance assessments of a repository
	Model validation (MV)	Direct application in model development and validation for safety/performance assessments of a repository
Qualitative	Scenario development including FEPs (SD)	Indirect application as FEPs and scenario developments necessary for safety/performance assessment of a repository
	Understanding improvement (UI)	Direct application as information for understanding improvement on the behaviors of disposal system including EBS and NBS
	Demonstration and education (DE)	Direct application as demonstration and education materials to the public and stakeholders for the radioactive waste disposal

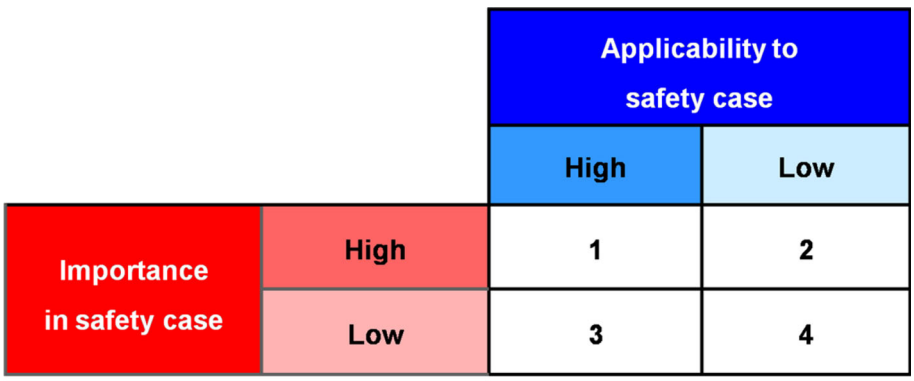
The abbreviations (codes) of the different categories are inserted in column 11 of the spreadsheet (Fig. 3)

including FEPs), UI (Understanding Improvement), and DE (Demonstration and Education). Table 2 shows the categories for the methods used to apply the natural analogue information to the safety case, and explains the category codes used in the natural analogue database.

3.3 Suitability for the Korean safety case

The most important thing to be considered in the application of natural analogue information to the safety case is probably the determination of what natural analogue information is applicable to a specific safety case, namely the proposed deep disposal of high-level radioactive waste in South Korea. The collected natural analogue data were classified into four categories based on their importance in the safety case and their applicability to a specific safety case. Figure 4 shows the classification categories and criteria of the natural analogue datasets used to evaluate the suitability of natural analogue information to the safety case.

During the review process, several experts familiar with radioactive waste disposal participated in the evaluation and categorization of the natural analogue datasets. Based upon the evaluation of suitability of natural analogue information to the Korean safety case, 176 natural analogue datasets were evaluated and categorized in terms of four levels of suitability, suitability levels SL-1–SL-4 (Fig. 4). Among these, 75 were categorized as SL-1 (natural analogue data important in a safety case, and with high applicability); 63 as SL-2 (natural analogue data important in a safety case, but with low applicability); 15 as SL-3 (natural analogue data not important in a safety case, but with high applicability); and 23 as SL-4 (natural analogue data not important in the safety case, and with low applicability).



Categories	Description
1 (SL-1)	NA information important in the safety case with high applicability
2 (SL-2)	NA information important in the safety case with low applicability
3 (SL-3)	NA information not important in the safety case with high applicability
4 (SL-4)	NA information not important in the safety case with low applicability

Fig. 4 Classification of natural analogue information data based on suitability for application to the South Korean safety case. *SL* suitability level: the numbers *1–4* represent different levels, from *high* (*1*) to *low* (*4*), and are inserted in column 12 in the spreadsheet (Fig. 3)

Table 3 Natural analogue information categorized as SL-1 (suitability level 1) based on the evaluation of suitability for the Korean safety case

System	Components/processes	Application method	Properties/phenomena
EBS	Spent fuel	MV	Dissolution rate and processes of spent fuel
		MV	Radiolysis effect on dissolution of spent fuel
		DE	Criticality of spent fuel
		UI	Formation and alteration of uranium minerals
	Metal	ISP, MV, DE	Corrosion rate and corrosion products of copper
		UI, DE	Durability and longevity of copper
		ISP	Pitting factor of copper
		MV, DE	Durability and longevity of iron
		MV, UI, DE	Corrosion rate of iron
		UI	Radionuclide retardation by corrosion products of iron
	Bentonite	UI	Colloid filtration by bentonite
		DE	Hydraulic barrier of bentonite
		DE, UI	Interactions of bentonite with other materials
		DE	Durability and longevity of bentonite
		SD, UI	Alteration of bentonite
		UI	Thermal alteration of bentonite
	Concrete and cement	MV, UI	Interaction of cement with groundwater
		UI	Interaction of cement with rocks
		DE	Durability and longevity of cement and concrete
Host rock	Elemental solubility and speciation	MV	Calculation and measurement of solubility and speciation of RNs in groundwater
	Elemental retardation	UI	Radionuclide mobility in rocks
		UI	Radionuclide migration and retardation through rock fracture
		UI	Fracture filling and coating materials
		UI	Interactions of RNs with minerals/oxides
		MV, SD	Migration model development and validation
		ISP, UI	Determination of distribution coefficient
	Matrix diffusion	ISP, MV, UI	Rock matrix diffusion depths
		ISP, MV, UI	Rock matrix diffusion coefficients
	Radiolysis	UI	Effects of radiolysis in the groundwater
		MV, UI	Gas generation rate by radiolysis
	Redox front	UI	Migration and retardation in the redox front
		MV, UI	Migration of redox front
	Colloids	ISP, UI	Characteristics of aquatic colloids
		ISP, UI	Radionuclide uptake by colloids
		ISP, UI	RN migration by colloids
	Microbial activity	UI	Characteristics of microbes
		UI	Migration and retardation of RNs by microbes
	Gas generation and migration	UI	Gas migration and reactions with the geosphere
		ISP, UI	Gas generation rate

For explanation of the abbreviations in the "Application method" column, see Table 2

For example, the natural analogue data for rock matrix diffusion depths of radionuclides in granite were considered SL-1, because the rock matrix diffusion depth is very important for the safety case, as well as being highly applicable to the Korean safety case. The importance of rock matrix diffusion lies in fact that it provides a mechanism for enlarging the rock surface in contact with the migrating radionuclides from fracture surfaces to a portion of the bulk rock. The data on the distribution coefficients of radionuclides obtained from the Maqarin natural analogue site in Jordan were considered SL-2, because the distribution coefficients of radionuclides in highly alkaline groundwater

are nearly applicable to the Korean safety case, while the radionuclide behavior in a highly alkaline groundwater, reacting with cement materials, are important in the safety cases of most countries. The natural analogue data for bio-colloid transport were categorized as SL-3 since bio-colloid transport has not been considered to be important in safety cases, although these data could be highly applicable to the Korean safety case. Finally, the natural analogue data for radionuclide retardation by secondary alteration products of silicate glass were categorized as SL-4, because silicate glass is not under consideration in Korea as a waste form.

Table 3 lists all of the selected SL-1 natural analogue information which is important and which will be considered in the Korean safety case. This information will provide insights and perspectives in planning RD&D (Research, Development, and Demonstration), including natural analogue studies, for radioactive waste disposal in Korea. In addition, this information will guide the inclusion and utilization of natural analogue information in the safety case.

4 Conclusions

The status of natural analogue studies in Korea was briefly summarized. A natural analogue database was collected and developed for effective application of natural analogue information to the safety case. In the database, the collected data were categorized according to the components or processes of EBS and host rock, and their application methods. Natural analogue data were also evaluated based on suitability for the Korean safety case. Among 176 natural analogue datasets collected, 75 were selected as important and highly applicable to the Korean safety case. The results from this study will be used to provide necessary information to develop the safety case for radioactive waste disposal and to serve as a regulation guide for the inclusion of natural analogues in the development of safety case. The natural analogue database will be further updated by collecting and evaluating natural analogue data published in the open literature.

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